Spin and charge transport in graphene devices in the classical and quantum regimes

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Chapter 1

Introduction

Abstract

In the past decade, the discovery of new materials and phenomena lead to a boost in research and applications of spin-based devices. Graphene, a one atom thick graphite layer, already holds many world records such as highest electronic mobility at room temperature and hardest known material. Furthermore, graphene is considered as one of the most promising material for spintronic applications due to predictions that spins can retain their information for very long times and carry this information for large distances. However, first experiments performed on graphene spin-based devices showed spin lifetimes orders of magnitude smaller than the initial estimates. Since then, efforts in both experimental and theoretical fronts have been done to find the culprits of this discrepancy between the experimental results and the initial theoretical predictions. Due to its high electronic quality, graphene can show quantum behaviour at larger length scales than most metals and semiconductors. This makes it easier to study the effects of quantum confinement and quantum interference in graphene-based devices.

In this thesis I present my research on charge and spin transport performed in the past 4 years. In the first three introductory chapters I present graphene's electronic and spintronic properties, and also the experimental methods used for my research. Motivated by previous experimental results for ballistic graphene nanoconstrictions obtained in our group and presented in chapter 2, we studied theoretically the electronic transport as a function of magnetic field in one-dimensional graphene structures, such as ribbons and constrictions of different shapes. Next I present our experimental work done on spin injection and transport in graphene nanostructures showing that these spin-based nanodevices have good prospects for spintronics, but contact induced spin relaxation can be stronger than on regular graphene spin valves. Chapter 7 shows our work done in spin dependent quantum interference in graphene, showing that the spin signal can be modulated by orders of magnitude by the application of gate voltages. In the pursuit of the study of spin relaxation in pristine graphene devices we studied the spin transport in high quality suspended graphene devices, chapter 8, showing that the nonsuspended regions dominate the measured spin relaxation. In chapter 9 I present the work performed in hexagonal BN encapsulated graphene in which, due to a double gated structure, the carrier density and the electric field could be controlled separately. We show that increasing the transverse electric field results in the increase of Rashba-type spin orbit fields pointing preferentially on the graphene plane. In the last chapter I give an overview of all the results presented here and conclude with a short outlook on graphene spintronics.

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1.1 Spintronics

In addition to their mass and charge, electrons also possess an intrinsic magnetic moment called spin. Spin is a purely quantum mechanical property of particles with no classical analogue. Electrons have a spin of $\hbar/2$ and therefore have two eigenstates, often labelled as *spin up* and *spin down*.

Analogous to the electronic charge in electronics, the spin can also be used to convey and store information. This is known as *spintronics*. The first attempts to apply spintronic concepts in devices was done by using the effect known as Giant Magneto-Resistance (GMR) discovered by the groups of Albert Fert and Peter Grünberg in the end of the 1980's [1, 2]. Soon after its discovery, GMR was already applied in computer hard disks, and a similar effect known as Tunneling Magneto-Resistance (TMR) is applied in hard disks today.

The basic idea of the GMR effect is that a heterostructure composed of two ferromagnetic materials separated by a non-magnetic layer shows a difference in resistance if the magnetization of the layers point parallel or antiparallel to each other. This results in a high resistance state and a low resistance state, which are interpreted by the computer as bits 0 and 1.

Recent spintronic applications include the spin transfer torque random access memory devices (STT-RAM) and magnetic random access memory (M-RAM) which might replace the current electric based RAM, leading to faster booting and lower power consumption since the magnetic based RAMs are nonvolatile. Other concepts are also been heavily studied. The field of quantum information and computation [3], for example, uses the fact that spins can carry a large amount of information per bit since their direction is mapped on the Bloch sphere. Therefore they can be used as quantum-bits (qubits). In addition to that, spin states can be entangled. This leads to a completely different paradigm for computation, which is used in a machine proposed as a quantum computer. The quantum computation approach reduces enormously the computation time for difficult tasks.

A less complex and more direct application in the CMOS industry is the use of spins as classical information carriers which is represented by the proposal of the Datta-Das spin transistor [4]. In order to obtain such a device we have to have three essential tasks: spin injection, manipulation and detection. In the original Datta-Das proposal, such device consists of a semiconductor with a large spin-orbit interaction, which allows for electrical manipulation of the spin, while the injection and detection are done using ferromagnetic contacts. However, there are two main problems with this proposal. First, a semiconductor is usually highly resistive which makes the electrical spin injection a difficult task [5, 6]. Second, the spin information lifetime in semiconductors with high spin-orbit interaction is usually too low, leading to loss of spin information before the manipulation can be performed. To solve the first problem a lot of attention has been given into the study of the improvement of electrical spin injection into semiconductors either via DC methods using ferromagnetic electrodes and cleverly engineered interfaces [7–11], or by an AC approach in which spins are dynamically injected into the semiconductor by oscillating the magnetization of a ferromagnet [12]. In order to achieve long spin lifetimes several materials have been studied. And one material that stands out as a very promising material for spintronic applications is graphene, for the reasons explained below.

1.2 Graphene

Graphene [13] is an all-carbon material with the atoms arranged in a sp^2 hybridization in a honeycomb lattice in a 2-dimensional (2D) version of graphite. In 2004 the demonstration of an easy method for isolation of graphene flakes [14], known as the scotch-tape method, lead to a boost in the research of the physical and chemical properties of graphene. Graphene has already shown to have several outstanding qualities such as its high mechanical strength [15], high thermal conduction [16], and optical properties [17].

But perhaps the most outstanding characteristic of graphene is its high electronic mobility even in low-quality devices at room temperature [18, 19]. This is due to two reasons: it is difficult to backscatter electrons in graphene [20], and the electron-phonon coupling in graphene is relatively low [21]. For this reason graphene has attracted a lot of attention for electronics, with applications ranging from high-frequency [22] to transparent touch screens [23]. The high electronic mobility of graphene leads to a long carrier mean free path which results in easier fabrication of devices that explore the quantum nature of the carriers such as quantum dots [24, 25], quantum interference devices [26, 27] and the observation of (fractional) quantum Hall effect [18, 28, 29].

Graphene has already shown great potential for applications in electronics. However, the lack of a bandgap results in transistors with poor on-off ratios which strongly limits the application of graphene in digital electronics. Perhaps the most probable application in electronic components will be in high-speed electronics [30], since those do not require high on-off ratios and the high mobility of graphene leads to very large cut-off frequencies ($>$ 100 GHz) [22].

Recently, it was demonstrated that the scotch-tape method can be also used to exfoliate other layered materials in order to obtain their 2D analogous [31]. Among these layered materials, transition metal dichalcogenides (TMDs) stand out as very promising for digital electronic applications. These materials have chemical composition of the type MX_2, where M is a transition metal and X a chalcogen element. When thinned to a single layer, TMDs show direct band gaps in the order of 1.5 eV, which results in large on/off ratios in the order of 10^8 with mobilities in the order of hundreds of cm^2/Vs [32].

In an effort to introduce graphene into the industry an European-wide project has been recently funded by the European Union. This project, called *Graphene Flagship*, involves researchers from several countries and performing research in several fronts. Also, a strong collaboration with industries is encouraged, strengthening the chances for future graphene applications. For that, a roadmap for graphene was developed [33] and aims to have the first applications available before the year 2020. It is important to point out that the Graphene Flagship not only opens the door for applications based on graphene but also for other layered materials, since the technology developed to fabricate graphene devices can be transferred to these other materials as exemplified by the scotch-tape technique. As mentioned before, graphene is a promising material for spintronic applications. Therefore, the Graphene Flagship also includes graphene spintronics as one of its work packages.

1.3 Graphene spintronics

The coupling between the orbital and spin angular momentum of the electron, the spin-orbit interaction [34], is a key point for the loss of spin information in semiconductors [35]. Another factor that can contribute to the loss of spin information is the hyperfine interaction [34], i.e. the interaction between the electronic spin with the spins from the nuclei. The spin-orbit interaction scales with the atomic weight as Z^4. Carbon, being a light element, shows a low spin-orbit coupling. This, combined with the fact that most of the carbon atoms (^{12}C) do not have spin, result in the prediction that graphene could maintain electronic spin information for long times [36]. For this reason graphene is a strong candidate for use in spintronic applications. Theoretical values for the intrinsic spin relaxation time in graphene range from τ_s=0.1 - 10 μs and the theoretical values for the spin relaxation length is in the order of $\lambda_s \approx$ 100 μm [36–38]. Although the first experiments to measure τ_s and λ_s showed values a few orders of magnitude lower than those previously predicted, with $\tau_s \approx$ 0.1 - 0.3 ns and $\lambda_s \approx$ 1 - 3 μm [39–42], most recent experiments using pure spin currents have been closing in on the predictions, with values of $\tau_s \approx$ 0.4 - 2.3 ns and $\lambda_s \approx$ 4 - 12 μm [11, 43–49]. Recent measurements based on few graphene layers on a SiC substrate at low temperatures using 2 probe devices in which spin effects are superimposed on the charge transport estimated $\lambda_s \approx$ 100 μm and $\tau_s \approx$ 100 ns [50]. However, it is important to notice that many different effects that can mimic spin signals, such as the magneto-coulomb effect, and could not be excluded by these experiments.

There is still a heavy debate, both in experimental and theoretical grounds, to understand what are the limiting factors for spin transport in graphene devices. However, even though the theoretical limits for graphene spintronics are still not reached, graphene already stands out as the best known material to host and transport spins at room temperature. The values for longest spin relaxation time and length at room temperature were measured in graphene based devices with $\tau_s \approx 2$ ns and $\lambda_s \approx 12$ μm [48, 49]. Graphene also showed transport of spin information over 20 μm at room temperature [46], the longest distance reached so-far at room temperature.

The main questions in the field of graphene spintronics at the moment are:

1 - *What is limiting the experimental values for τ_s? Is it an intrinsic limit of graphene or is it due to impurities and adsorbates?*

The puzzling discrepancy between experimental and theoretical results has attracted the attention of most researchers in the field due to its importance on the understanding of spintronics in graphene and its limits [51]. Up to now the most probable explanation of such discrepancy is that impurities strongly affect the spin lifetime in graphene. It has been theoretically demonstrated that adatoms can locally change the spin-orbit fields leading to spin scattering [37, 52, 53]. Furthermore, a theoretical model also showed that even a small amount of magnetic impurities can cause a strong decrease in the measured spin relaxation time [54]. Experimental efforts to answer this question lead to the research of tunable mobility graphene spin valves [55] and ultimately, the study of the spin transport in devices with a very low amount of scatterers [45, 46, 48, 49].

2 - *Is it possible to manipulate spin electrically in graphene? Is it possible to have a Datta-Das type of transistor based on graphene?*

Although it is an essential step towards applications, the electrical manipulation of spins in graphene has received little attention. Since graphene has a very low spin-orbit interaction, the electrical manipulation of spins requires high electric fields. This can be obtained using high-κ dielectrics or dielectrics with high breakdown voltages. Furthermore, as it will be discussed in chapter 3, the study of the spin transport in combination with transverse electric fields gives information on the spin-orbit coupling in graphene. This information can be used to understand the limiting factors of spin transport in graphene, and therefore helping to answer question 1.

3 - *Is it possible to combine other qualities of graphene (mechanical, electrical and chemical) with spin transport? How do mechanical vibrations, strain and chemical doping affect the spin transport?*

The combination of the different qualities of graphene with spin transport has already generated very elegant papers, as for example the study of paramagnetic moments in graphene using pure spin currents [56]. In addition to that, since curvature effects in graphene result in extra spin-orbit terms [57], it would be interesting to study how the mechanical properties of graphene could be used to manipulate spins. More interestingly, quantum mechanical phenomena can be used to enhance and strongly modulate the spin signal. Carbon nanotube quantum dots showed the possibility of strong modulation and inversion of the spin signal [58, 59]. In graphene, Fabry-Perot cavities showed that quantum interference can also modulate the spin signal in quantum coherent devices [60]. A later work showed that using the universal conductance fluctuations phenomena in high magnetic fields makes it possible to study a spin resolved quantum interference in graphene [61]. Universal conductance fluctuations and weak localization measurements can also give relevant information on the spin relaxation mechanisms and spin-orbit fields involved [62, 63].

4 - *What is the best method to fabricate spin polarized contacts in graphene, and how much do they affect the spin transport? Is the experimental limit on τ_s due to contact induced spin relaxation?*

The effect of the contacts and interface resistance in the spin transport in graphene is a point that has been studied extensively in the past years [11, 41, 50, 64]. It is known that, due to the conductivity mismatch problem [5], the spin injection and measured spin lifetime can be strongly affected for low resistive contacts [64]. However, contact resistance alone cannot explain the discrepancy of some experimental results [11, 50, 64], but the properties of the contact/graphene interface (e.g. roughness, dangling bonds and localized states) seem to also play a role on the measurements.

5 - *What happens to the spin transport characteristics when the number of graphene layers is increased to two, three or more? Does the charge screening of adding more layers help the spin transport? Is the mechanism for spin relaxation different when more layers are added?*

The electronic properties of graphene (e.g. it's band structure) changes dramatically when more layers are added. While single layer graphene has a linear electronic dispersion, bilayer graphene shows a parabolic dispersion, and trilayer graphene has parabolic and linear dispersions superimposed. In addition to that, as more and more layers are added, the graphene layers away from the surface get electronically protected from the environment due to screening. Some work has been done towards studying the differences on the spin transport as a function of the number of layers [43, 44, 65]. These works have shown that the addition of more graphene layers seems to have a positive effect on the spin transport, increasing the spin relaxation time. Furthermore, the most relevant spin relaxation mechanism in bilayer graphene seems to be different from the one in single layer graphene, which is rather surprising.

This thesis focuses on the first four questions as described below.

1.4 This thesis

As stated before, this thesis focuses on the spin transport in graphene devices and tries to explore different aspects of it. I also give a short introduction to graphene, its electronic and spintronic properties, and the experimental methods used in this thesis.

The introductory chapters have the goal of explaining the relevant topics for the understanding of this thesis assuming that the reader has at least a graduate level in physics. However, sometimes the reader might find necessary to have the topics explained by a different point of view, or get more information about it. For this I selected from the references texts that provide a different explanation or more in depth information about the topics. This selection can be found in the *Supplementary literature* section at the end of each introductory chapter.

A brief summary of each of the following chapters is presented below.

Chapter 2 introduces the basic concepts of graphene and electronic transport such as graphene's crystal lattice and band structure, graphene field-effect transistors, quantum Hall effect and conductance quantization due to confinement.

Chapter 3 introduces basic concepts of spintronics and spin transport, and later focuses on the special case of graphene. The methods for electrical spin injection / detection in graphene, the conductivity mismatch problem, Hanle effect, and spin transport in inhomogeneous systems are discussed. At the end of the chapter I give a thorough overview of the current understanding and models for spin relaxation in graphene and the effect of an electric field on the spin relaxation in graphene.

Chapter 4 presents the experimental techniques used throughout this thesis. The sample fabrication for the different types of samples used here, the experimental setup and the electrical measurements are carefully described.

Chapter 5 presents a theoretical work on the transition between the conductance quantization due to geometrical confinement to the quantum Hall effect in graphene nanoconstrictions and nanoribbons. The effects that different shapes of the constrictions have on the electric transport properties are discussed and we compare the results obtained here with previous experiments performed in our group, which are used as example for the effects treated in chapter 2.

Chapter 6 shows the study of spin transport and accumulation in different graphene nanostructures with dimensions smaller than the spin relaxation length. Here we try to understand the role of the edges and confinement on the spin transport in graphene. We apply the model for spin diffusion to explain our experimental results.

Chapter 7 demonstrates the effect of quantum interference in a quantum coherent nonlocal graphene spin valve. We show that by changing the interference pattern in the device, the nonlocal spin signal can be strongly modulated and even reverse polarity. This indicate that quantum mechanical effects can be used in combination with spin transport in graphene to obtain novel type of devices.

Chapter 8 presents the first study of the spin transport in high quality suspended graphene spin valves. Here we try to understand the effect of the environment on the spin transport in graphene by removing the substrate and most of impurities and adsorbates. We give a lower bound for both the spin relaxation time and length. By a theoretical model we explain our results and show that the measured spin relaxation time is limited by the nonsuspended (lower quality) regions.

Chapter 9 shows the study of the spin transport in hBN encapsulated spin valves.

It is demonstrated that, by isolating the graphene flake from the environment using non-invasive hBN flakes, the spin transport in graphene is considerably improved. We achieve the highest spin relaxation length and times at room temperatures reaching 2 ns and 12 μm respectively. The use of a double gated geometry further permits us the study of the effect of an electric field in the spin relaxation, which allows for the control of the spin relaxation in graphene by Rashba-type spin orbit fields. This study also gives insights on the origins of the spin orbit fields in the absence of electric fields.

Chapter 10 gives a general conclusion for this thesis and a brief outlook on interesting questions and new paths for further research.

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